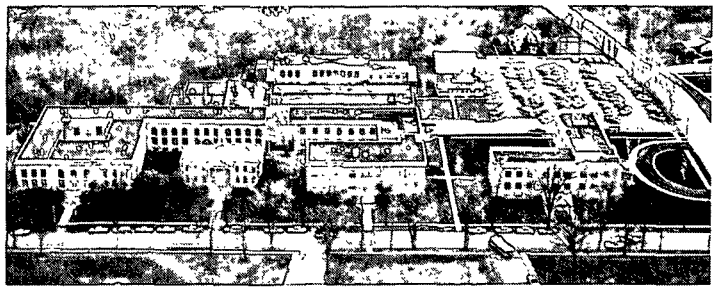

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THE INSIDE-OUT TECHNIQUE FOR NUMERICAL INTEGRATION — ILLUSTRATIONS AND APPLICATIONS

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The Inside-Out Technique for Numerical Integration - Illustrations and Applications

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THE INSIDE-OUT TECHNIQUE FOR NUMERICAL INTEGRATION - ILLUSTRATIONS AND APPLICATIONS

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ABSTRACT

Applications of the Inside-Out Technique are presented illustrating the enhanced speed, stability, and accuracy over integration methods such as the variable-step predictor corrector method of Adams-Bashford or Adams-Moulton.

INTRODUCTION

A variety of processes in pulping and bleaching occur in plug-flow chemical reactors. Examples include bleaching and extraction such as chlorination, peroxide bleaching, and alkaline extraction. Conservation equations on the chemical species in the reactor result in one or more ordinary differential equations describing the change in chemical concentration of each species with position or reactor space-time. Steady-state simulation requires the repeated integration of these equations. Such repeated integrations can add significantly to the time required for convergence (close of mass balances) and in some situations result in very poor convergence characteristics.

Bleaching reactions are characterized by a rapid initial rate followed by a very slow "tail." The reactions tend to be second-order or higher in the key reactant, either lignin or absorption coefficient. Another important consideration is that in process simulation, we are not concerned about detailed intrareactor profiles but only with the outlet concentrations and the chemical consumptions and overall level of brightening. Ideally, we seek a technique that will compute the exit concentrations in the shortest computation time.

Conventional techniques such as Euler, Trapezoidal Rule, variable step, predictor corrector, and Runge-Kutta methods all compute the dependent variables (chemical species concentrations) by integrating forward in time for a fixed or variable time step. However, for the reactions of interest, the time step must be very small at the beginning and very long at the end. There is no general way to vary the time step and still preserve the accuracy of the solution.

MAPPS, the process simulation package developed at the Institute, incorporates a novel technique for the integration of the kinetics for bleaching and extraction reactions which overcomes the difficulties with the conventional techniques. The essence of the technique is as follows: Instead of varying time and computing the species concentrations, we vary the concentration of the key component and compute the secondary component concentrations and time. We then check for limiting conditions such as consumption of secondary chemicals or elapsed time.

Because the roles of independent and dependent variables are reversed, the technique is referred to as the Inside-Out or I-O method. The technique has the following features:

1. The key reactant is usually the highest order reactant and the one in which the reaction stoichiometry is written.
2. Concentration of the key reactant is bounded so the incremental change in the key reactant, ΔC_a, is some fraction of the maximum. The step size has a natural value depending on the desired coarseness of the integration. For example, if the inlet concentration of A is 1 and ten steps are desired, the step size is 0.1.

3. The stoichiometric relations are such that the equations for the remaining reactants are either algebraic or can be reduced to algebraic relations. Thus, the concentrations of the secondary components, B, C, D, etc., can be readily computed over each increment of A. In fact, the procedure allows for highly accurate estimates of B, C, etc., by evaluating the stoichiometric relations at the half interval of C_a.

4. This brings up another special advantage of the method. Since the material balance equations for the steady-state plug flow reactor are of the following general form

$$\frac{dC_i}{dt} = -R_i\ (C_1, \ldots, C_i \ldots C_N)\quad i = 1,M$$

subject to initial condition and general stoichiometric relations

$$t = 0 \qquad C_i = C_i^0$$

$$g_i\ (C_1, \ldots, C_i, \ldots C_n) = 0 \quad j = 1,L$$

$$N = L + M$$

R_i can be accurately approximated over each increment dC_a by the half-interval values of A, B, C, etc.

The I-O method takes on the following general form

$$dt = \frac{-dC_i}{R_i}$$

For R of a general form, the finite-difference version of the I-O method must be used. For a single reactant, A, we have

$$\frac{dC_a}{dt} = -R(C_a)$$

$$\Delta t^i = t^{i+1} - t^i = \frac{-\Delta C_a{}^i}{R} = \frac{C_a{}^i - C_a{}^{i+1}}{R}$$

$$R = R\,(C_a{}^{i+1/2})$$

When R has the form of a power law as shown below, the semianalytic version of the I-O method can be used. In this latter version, all the terms in R except that involving C_a are lumped into the rate constant. The expression is then integrated exactly for C_a and the equation is solved for dt.

$$R = kC_a{}^P C_b{}^q C_c{}^r \ldots C_n{}^z$$

$$\Delta C_b{}^i = g_b(C_a{}^i) - g_b(C_a{}^{i+1})$$

$$\Delta C_c{}^i = g_c(C_a{}^i) - g_c(C_a{}^{i+1})$$

Let

$$K^i = k\overline{C_b}{}^i q \overline{C_c}{}^i r$$

$$\overline{C}_b{}^i = C_b{}^{i-1} + \Delta C_b{}^i/2$$

$$\overline{C}_c{}^i = C_c{}^{i-1} + \Delta C_c{}^i/2$$

The semianalytic solution is

$$p \neq 1 \quad C_a{}^0 \neq 0$$

$$\Delta t^i = \{1/C_a{}^{i+1(p-1)} - 1/C_a{}^{i(p-1)}\}/K'^i$$

$$K'^i = (p-1)K^i$$

For $p = 1$ and $C_a{}^0 = 0$ other forms apply.

Special forms of the equation are required for first-order reaction in A and for zero initial concentration in A. This background material is discussed in more detail in a previous report (1).

When time exceeds the maximum allowed reaction time or one of the secondary components is consumed, the I-O method backs up and interpolates to the correct outlet value of the key reactant.

The advantages of the techniques are increased stability, accuracy, speed (fewer iterations and fewer functional evaluations) and also increased flexibility. The latter refers to the reduced number of parameters needed to specify the integration compared to a variety of other methods. Other more subtle advantages could also exist.

The disadvantages are that although the method works well for the limited class of first order nonlinear differential and algebraic equations, it is not clear whether the technique can be generalized to any system of equations. For example, if the equation defining Y is of the form,

$$dY/dt = F(Y, t)$$

the I-O technique becomes implicit in t and time must be solved iteratively at each time step. There may not be any particular advantage to doing this over the conventional techniques. Another example is the case where Y has a series of extrema such as an oscillatory function. Then the I-O technique would require checking for a change in sign of Δt at each step. This may reduce its usefulness in such situations.

OBJECTIVE

We wish to quantify the improvement of the I-O technique over the more conventional but powerful variable-step predictor corrector technique of Adams-Moulton as found in the DGEAR package of the IMSL library. This method will be referred to simply as Gear's method in the following discussion. Also in the discussion, the finite-difference version of I-O will be referred to as FDIO and the semianalytic version of I-O as SAIO.

DISCUSSION

Five example problems were set up to compare the two methods. Comparisons are made on the basis of whether or not the methods failed, the accuracy of the answers and the speed of the solution.

The following applications were selected:

1. Case having an exact solution where Euler's method fails. We compare the absolute and relative error of FDIO with the analytic solution. In this case SAIO gives the exact solution directly.

Application 1

$$\frac{dy}{dt} = \sqrt{y} \quad y(0) = 0$$

2. No exact solution is evident so the finite-difference version of I-O is compared against Gear's method.

Application 2

$$\frac{dy}{dt} = \ln y \quad \begin{array}{ll} y(0) = 1 & \text{Case 1} \\ y(0) = 1.5 & \text{Case 2} \end{array}$$

3. Simplified chlorine bleaching. Nonlinear coupled ODE's.

Application 3

$$\frac{dXLF}{dt} = -2.06\ XLF^{1.1}\ XCl$$

$$\frac{dXCl}{dt} = 1.5\ \frac{dXLF}{dt}$$

$$XLF(0) = 0.06$$

$$XCl(0) = 0.002$$

4. Higher order reaction. Simplified alkaline extraction.

Application 4

$$\frac{dy}{dt} = -\ 100\ y^5\ z^{0.5}$$

$$\frac{dz}{dt} = 0.5 \frac{dy}{dt}$$

$$y(0) = 1$$

$$z(0) = 10$$

5. Extremely rapid reaction or extremely stiff system of two equations.

$$\frac{dy}{dt} = -k\ y^5\ z^{0.5}$$

$$\frac{dz}{dt} = 0.5 \frac{dy}{dt}$$

$$k = 100 + 1 \times 10^5\ e^{\ 1/y}$$

$$y(0) = 1$$

$$z(0) = 10$$

Results of Application 1

Plotted on Figure 1 are time on the vertical axis and y on the horizontal axis. The SAIO version predicts the exact solution shown on the upper curve. The FDIO version is shown on the lower curve for a value of Δy of 0.01. Euler's method fails for this problem for two reasons: 1. the initial slope is zero so the solution is "stuck" at the origin, and 2. the initial derivative of $y^{1/2}$ at $y = 0$ blows up so there is no way to evaluate the initial step. In fact all the derivatives of $y^{1/2}$ fail to exist at $y = 0$. Since I-O changes y instead of t, there is no problem with zero initial conditions or zero initial slopes with the I-O method.

Figure 1 here

Figure 2 shows the absolute and relative error of FDIO. The absolute error is defined as the difference between the two curves in Figure 1, while the

relative error is the difference divided by the value of time predicted by SAIO.

Figure 2 here

Figure 2 shows that instead of rising continuously as in other finite difference techniques, the absolute error remains constant after an initial increase. The absolute error can be made arbitrarily small by reducing the value of Δy (i.e., by increasing NY). This increases the total number of steps required. The relative error first increases from zero to its maximum after the first step and then asymptotically approaches zero. Thus the solution appears to be more accurate in relative terms rather than less accurate with more integration as are most other standard techniques.

Results of Application 2

In this case two situations are discussed. In the first case Gear's method fails to move from the initial condition because at $y(0) = 1$, $\ln(y0) = 0$ and the initial slope is zero. Thus Gear's method fails. The results of the FDIO are shown in Figure 3 for 10 increments of y. Again, time is plotted along the vertical axis and y along the horizontal axis since the roles of the variables are reversed. There is no way to compare the accuracy of this solution because there is no known analytical solution for this problem. However, the accuracy relative to that of Gear's technique can be shown for the second problem with a different initial condition, $y(0) = 1.5$, where Gear's method does provide a solution.

Figure 3 here

Figure 4 shows that FDIO with 10 increments determines precisely the same solution as Gear's method with approximately 100 Gear evaluations. It is not clear how many actual functional evaluations are required. For these

comparisons, the I-O methods were implemented on a PC AT while the IMSL routines were used on the Burroughs 6900. The actual run times appeared considerably longer for the Gear method than the I-O method even considering the differences in power and size of the machines.

Figure 4 here

Results of Application 3

The simplified chlorination material balances provide a comparison of the situation closer to the original motivation for the development of the I-O method. Results are summarized in Table I and Figure 5. Here we are primarily concerned with the outlet concentrations of Y and Z which are chlorine and lignin, respectively. Gear's method predicts that lignin is limiting, i.e., lignin is consumed. The reaction time is 45.5 seconds and the outlet concentration of chlorine is 0.0587. Gear requires 455 iterations to reach the solution.

Table I and Figure 5 here

Because of the power-law form of the rate, the SAIO version was used in this case. Four different values of NY were used to determine the sensitivity of the results to the number of increments. For NY of 10 and initial concentration of chlorine of 0.06, ΔXCl was 0.006. Note that the SAIO method arrived at the correct outlet concentration in only 2 steps for NY of 10. This resulted from the fact that all the lignin was consumed in the very first increment of chlorine from 0.06 to 0.054. There was one increment and an interpolation step counting as two steps. The outlet concentrations were correctly predicted but the reaction time was not. Although this was not important for the case of limiting Z, it could be important for the case of limiting time.

For this reason, NY was increased to 100, 500, and 1000 to determine at which level the predicted reaction time agreed with the Gear method. Between 500 and 1000 the two results agree. However, the actual number of steps was only 24 for the SAIO method compared to 455 for Gear's method.

Figure 5 shows the progress of the reaction time vs. y (chlorine concentration) for both techniques (lower curve) and the number of Gear functional evaluations (calls to the derivative functions) plotted on the upper curve. Note that the I-O method provides an internal solution as accurate as Gear even though this is not required of it. Each point on the lower curve represents a single I-O iteration while the number of Gear iterations to reach that point is shown in the upper dashed line.

Figure 5 here

Not shown is the fact that as the rate drops to zero as Z decreases, Gear's method actually fails for the parameters chosen. It appears that Gear needs special help exiting the integration on the flat portion of the curve when nothing much is happening. Because the I-O method increments Y rather than time, t, it forces the reduction of Z to below zero, thus forcing an interpolation to zero and an exit from the reactor.

Clearly the I-O approach is superior for this application. One would expect it to be even more effective for higher order reactions which should give Gear's method more trouble. This proves to be the case as shown in the remaining applications.

Results of Application 4

The speed of this reaction is shown in Figure 6. Practically no time has elapsed before y has decreased from the inlet value of 1 to 0.2. Here time is plotted on the vertical and y on the horizontal axis. In the alkaline extraction system y would represent dissolved lignin and z hydroxide ion. The two methods give essentially the same results as shown by the superposition of the two lines in the lower left portion of Figure 6. The number of Gear iterations is plotted on the upper line. In this situation, time is limiting. This is interesting considering the very high initial rates. Clearly, the rates drop to very low levels after the first second. Then almost 100 seconds are required to react the remaining 20% of y. Z is not limiting here.

Figure 6 here

The number of Gear evaluations reaches 116 and 10 calls to DGEAR were required compared to 10 total iterations for the I-O method. Also, five separate attempts were required with the Gear method to correctly specify the initial time step in order to reach a stable solution. This is not shown here.

Results of Application 5

This application was developed by adding a term to k in Example 4 to drive the rate to very high values as y approached zero. This is the opposite of the previous example in which the rate tended to decrease rapidly as y decreased. The results shown in Table II are surprising in that they show that time is again limiting. The SAIO version handles the very high rate with no problem while the Gear technique becomes oscillatory and eventually fails. The rate constant K* varies from 0.027 to 2.5×10^{47}. Failure occurs before the time limit is reached. Hundreds of iterations are used before the method fails.

With SAIO the solution is obtained smoothly in the same number of iterations as in Application 4. In this case total reactor space time is exceeded in the last step and interpolation yields the outlet values of y of 0.0008. Z is then determined from the stoichiometry. K', which is k multiplied by $z^{0.5}$, where z is evaluated at the half interval at each step, increases from 0.34×10^7 to 2700×10^7.

CONCLUSIONS

The I-O technique is highly versatile, very stable and extremely fast compared to the best available numerical integration routines. I-O is sufficiently general for most applications of interest such as reaction kinetics. More study will be required to extend this concept to the solution of general systems of ODE's.

ACKNOWLEDGMENTS

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1. Jones, Gary L. A new way of looking at the solution of kinetics expressions. IPC Technical Paper Series No. 266, Dec., 1987.

2. Numerical Methods, Software and Analysis: IMSL Reference Edition, McGraw-Hill, 1983:303-309.

NOMENCLATURE

x,y,z	dependent variables, chemical component concentrations
C_i	concentration of species i
g_i	stoichiometric or other algebraic relation on species i
t	time
R	reaction rate function
k	rate constant
K,K'	lumped rate constants
XLF	weight fractions fast lignin
XCl	weight fraction chlorine
NY	number of Y increments
I-O	Inside-Out method
SAIO	Semi-analytic I-O method
FDIO	Finite difference I-O method

Subscripts

a,b,c	species designation

Superscripts

0	initial value
i	ith increment
p,q,r...z	reaction orders

Figure captions

Figure 1. Application 1 - Solution by SAIO and FDIO methods for which Gear's method fails because of the nature of the initial condition.

Figure 2. Comparison of absolute and relative errors produced by FDIO for Application 1.

Figure 3. Application 2 (Case 1) - Solution predicted by FDIO for which Gear's method fails due to initial condition.

Figure 4. Superimposed solutions from FDIO and Gear for Application 2 Case 2.

Figure 5. Application 3 - Simplified bleaching kinetics showing superimposed solution from SAIO and Gear and number of Gear evaluations.

Figure 6. Application 4 - Superposition of SAIO and Gear and number of Gear evaluations.

TABLE I

APPLICATION 3
SIMPLIFIED CHLORINATION

I-O					GEAR			
NY	N	Yout	Zout	T	N	Yout	Zout	T
10	2	0.0587	0	14.5				
100	4	0.0587	0	28.9				
500	13	0.0587	0	43.1				
1000	24	0.0587	0	45.7	455	0.0587	0	45.5

TABLE II

APPLICATION 5
STIFF SYSTEM

I-O				GEAR		
Y	N	Time	K'*	N	Time	K*
1	1	0	0.34	19	0	0.027
0.9	2	0	0.38	6	0	0.031
0.8	3	0	0.44	7	0	0.035
0.7	4	0	0.52	6	0	0.041
0.6	5	0	0.66	6	0	0.051
0.5	6	0	0.92	7	0	0.076
0.4	7	0	1.5	6	0.00001	0.114
0.3	8	0	3.5	6	0.00001	0.247
0.2	9	0	18	6	0.00002	1.47
0.1	10	0.0001	2700	95	0.00002	22.7
0.0008	11	0.0001	2700	95	0.00002	2.5×10^{47}

*K or K' x 10^{-7}.

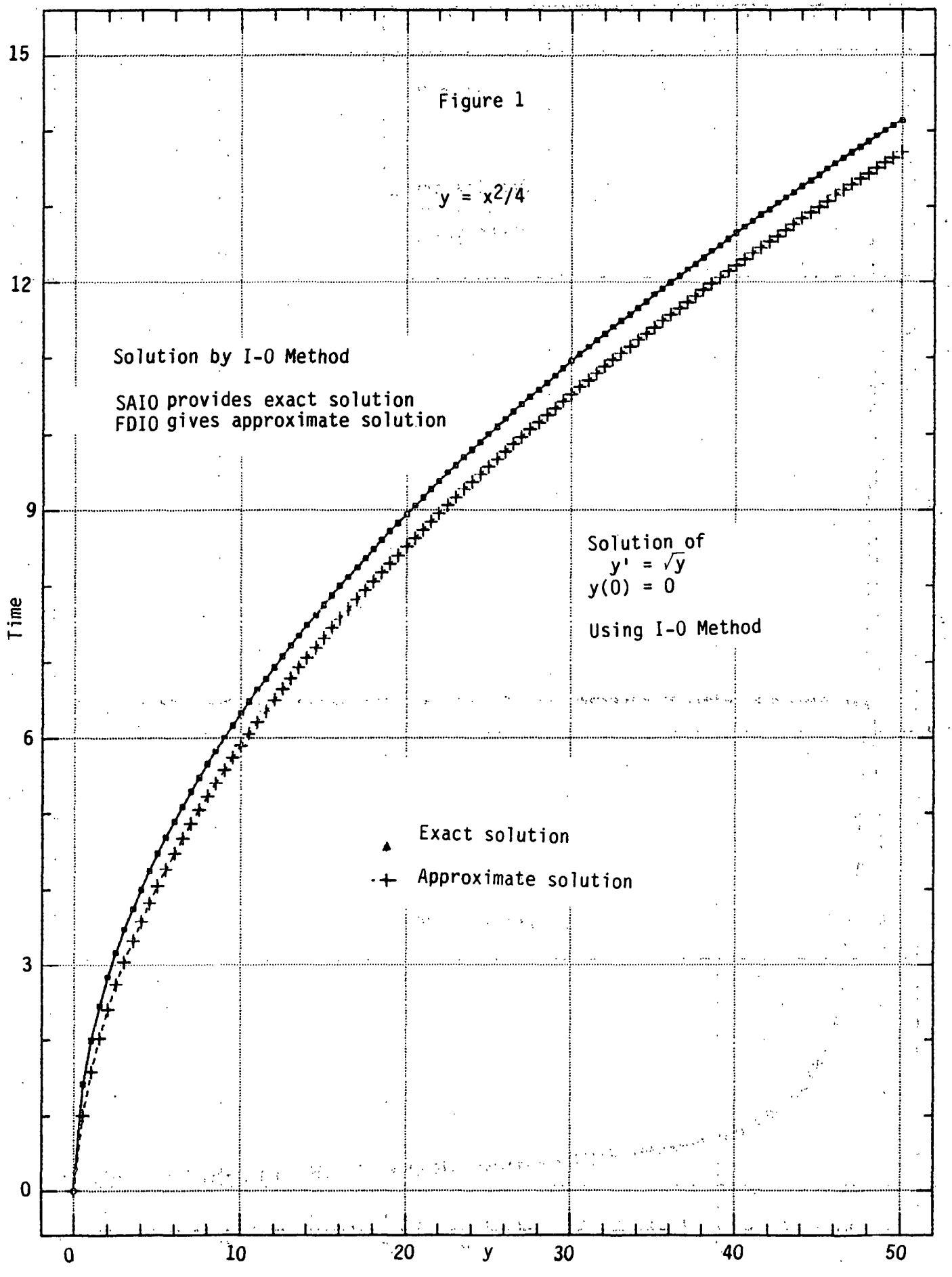
Figure 1
$y = x^2/4$
Solution by I-O Method
SAIO provides exact solution
FDIO gives approximate solution
Solution of
$y' = \sqrt{y}$
$y(0) = 0$
Using I-O Method
Exact solution
Approximate solution
Time
y
0
3
6
9
12
15
0
10
20
30
40
50

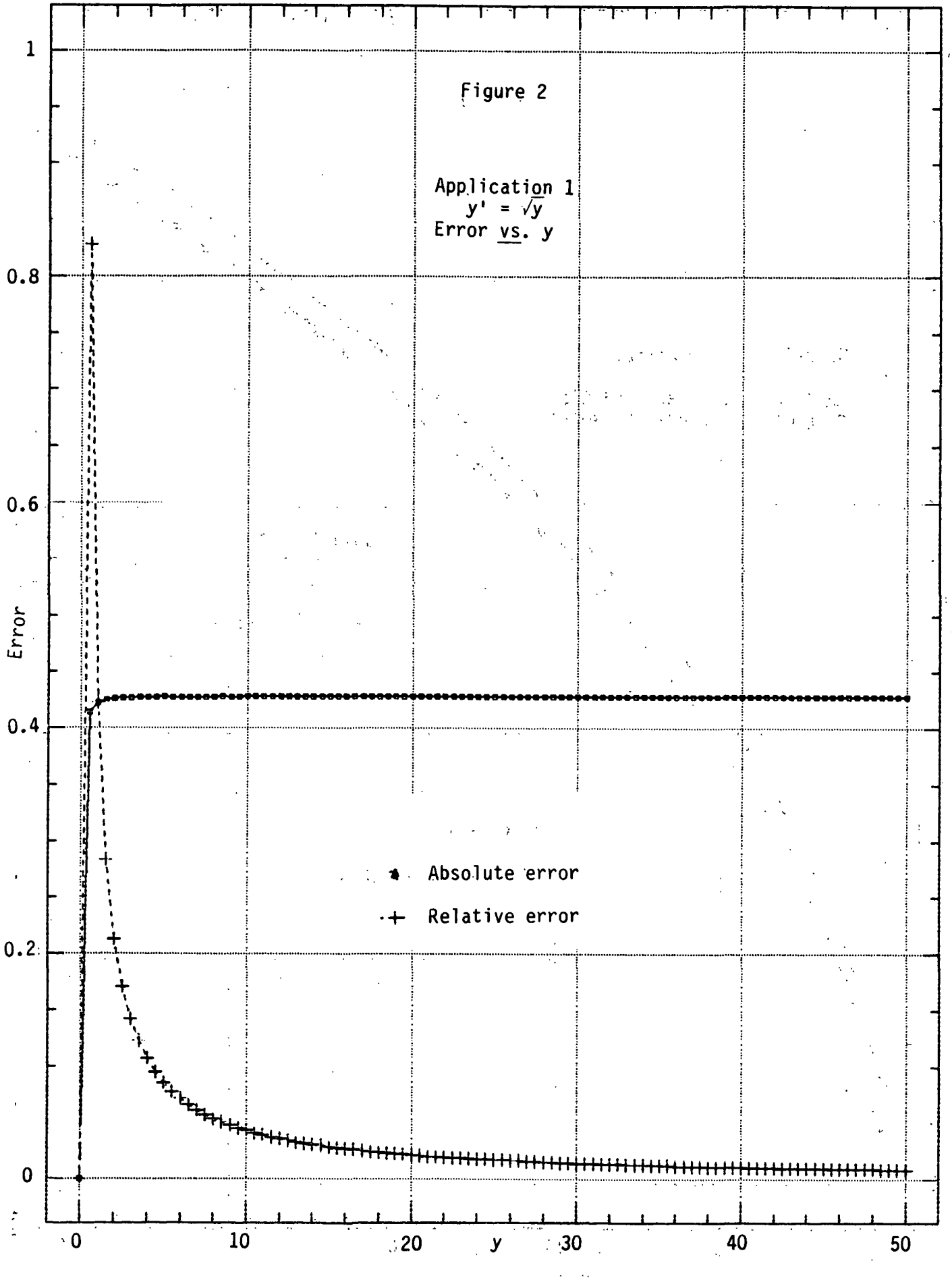
Figure 2
Application 1
y' = √y
Error vs. y
1
0.8
0.6
0.4
0.2
0
Error
Absolute error
Relative error
0
10
20
y
30
40
50

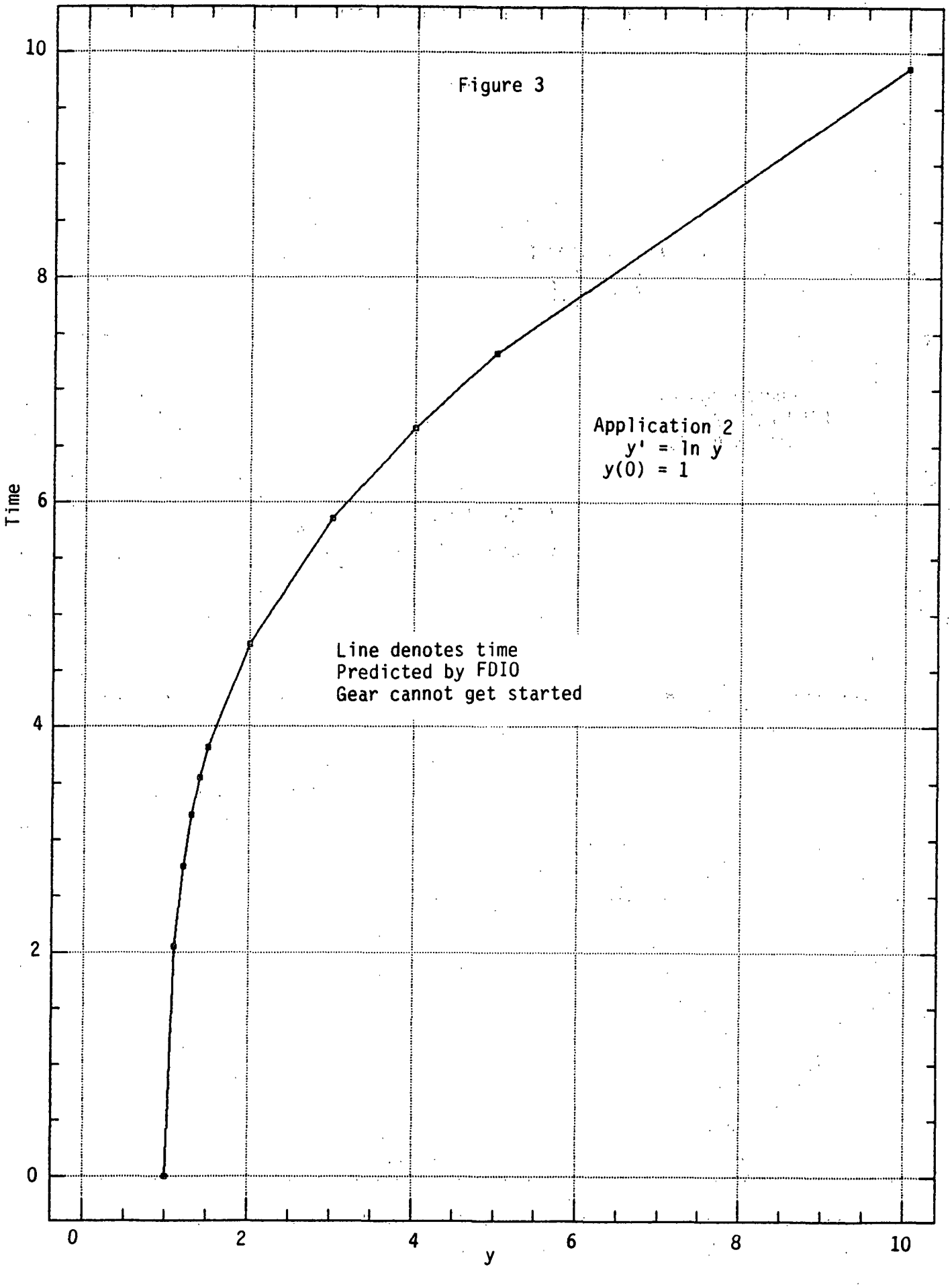
Figure 3
Application 2
y' = ln y
y(0) = 1
Line denotes time
Predicted by FDIO
Gear cannot get started
Time
y
0
2
4
6
8
10

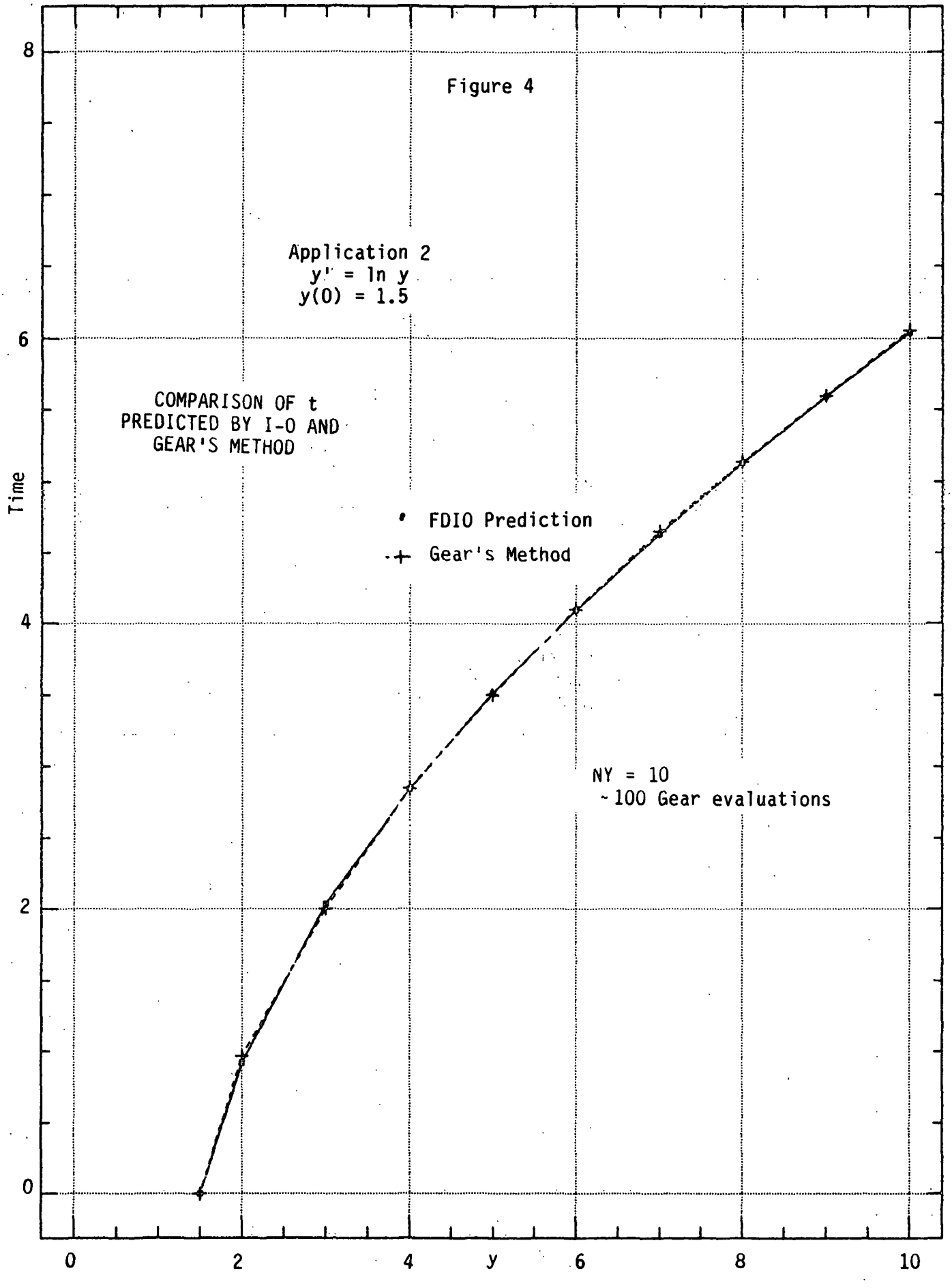
Figure 4
Application 2
y' = ln y
y(0) = 1.5
COMPARISON OF t
PREDICTED BY I-O AND
GEAR'S METHOD
FDIO Prediction
Gear's Method
NY = 10
~100 Gear evaluations
Time
y
0
2
4
6
8
10

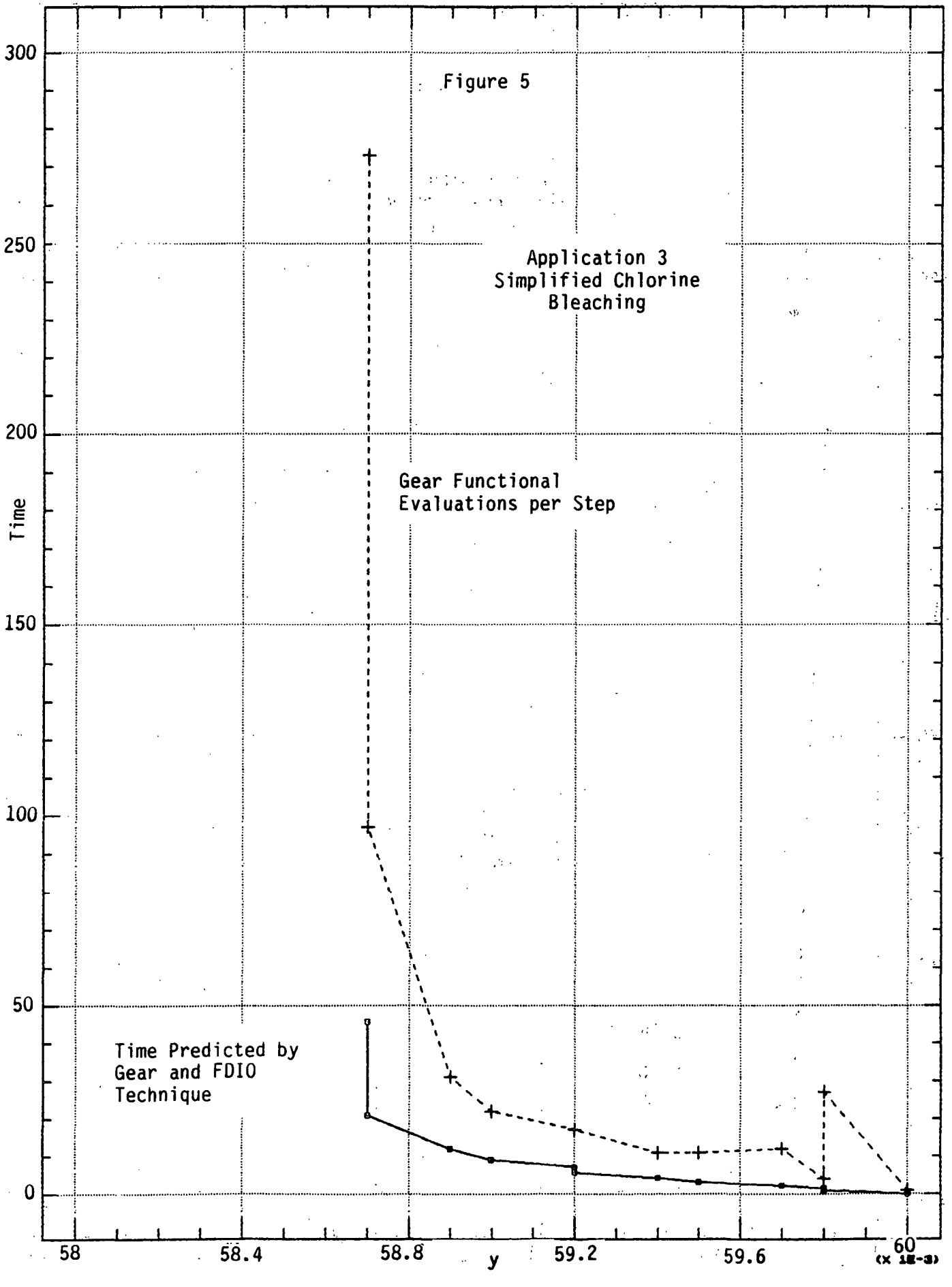
Figure 5
Application 3
Simplified Chlorine
Bleaching
Gear Functional
Evaluations per Step
Time Predicted by
Gear and FDIO
Technique
Time
300
250
200
150
100
50
0
58
58.4
58.8
59.2
59.6
60
y
(x 1E-3)

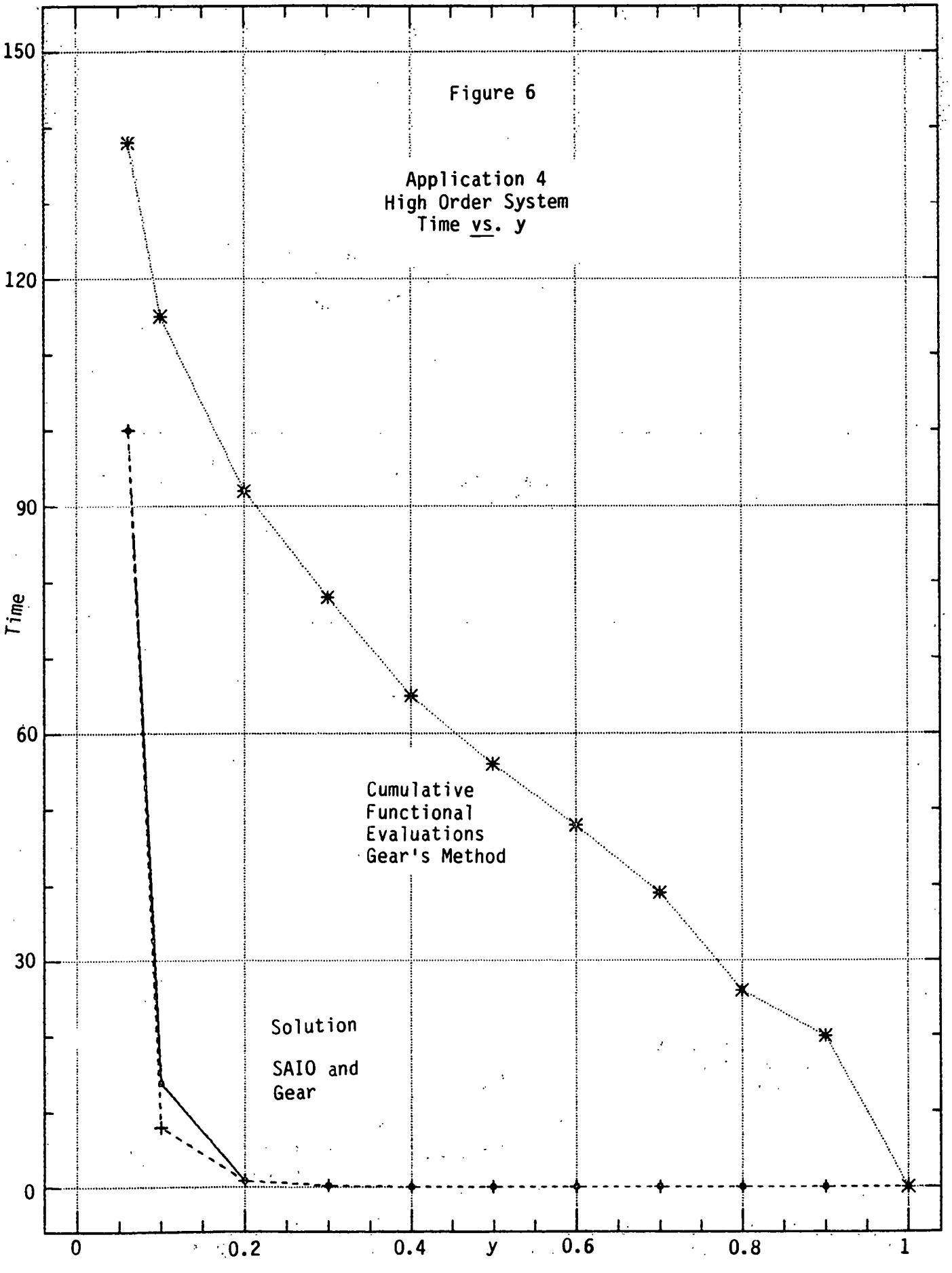
Figure 6
Application 4
High Order System
Time vs. y
150
120
90
60
30
0
Time
Cumulative
Functional
Evaluations
Gear's Method
Solution
SAIO and
Gear
0
0.2
0.4
y
0.6
0.8
1